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Micromechanics of creep and relaxation of wood. A review

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Abstract

Wood, like all polymeric materials, shows viscoelastic behaviour. The time dependent behaviour of wood depends on material anisotropy, temperature, moisture and stresses. To predict the behaviour of wood, numerous mathematical models have been developed largely relying on experimental results. In this paper, time dependent viscoelastic behaviour of wood is reviewed under constant and cyclic climatic conditions, separately. More emphasis is given on results obtained in recent years on the behaviour of thin wood tissues, single fibres, thermo-viscoelasticity of wood, influence of hemicelluloses and the modelling of the effect of transient moisture at the molecular level on the mechanical response.

Keywords: creep; hydrogen bonding; mechano-sorptive effect; micromechanics; modelling; relaxation; single wood fibre; viscoelasticity; wood tissue.

Introduction

Wood is a fibre composite consisting of various cell types and the cell wall is a composite biopolymer consisting of the polymers cellulose, hemicelluloses and lignin. Wood, like all polymeric materials, exhibits to a greater or lesser extent a viscoelastic behaviour (Ferry 1980; Morlier 1994). Its time dependent viscoelastic behaviour depends on the loading history, the temperature, the moisture-content (MC), MC history, as well as moisture variations. These parameters can interact and produce coupling effects. Owing to the large number of variables, any quantitative evaluation requires modelling (Hofstetter and Gamstedt 2008). A great deal of research has been carried out on wood viscoelasticity. In a collection of modelling, experimentation results of viscoelasticity are presented by Morlier (1994). The influence of MC and its variations on the creep was clearly illustrated by experiments on spruce (*Picea abies*) by Hoffmeyer (1990).

The viscoelasticity of wood and its modelling has been the object of many experimental and theoretical studies. Numerous data concerning the phenomenological (macroscopic) behaviour of wood under constant and variable climates is given in a review paper by Schniewind (1968). Dinwoodi et al. (1991) also reported a large number of experimental results on the viscoelasticity of wood and wood-derived materials. These results indicate that the creep depends mainly on the level of loading, the climatic conditions and wood species.

Numerous mathematical models have been developed for both linear and non-linear behaviour of wood (Morlier and Palka 1994). These models largely rely on experimental results and are established distinctly for constant and variable ambient conditions. This is probably due to the coupling effect that exists between transient moisture and mechanical stresses.

Thus, this paper presents separately the viscoelasticity of wood under constant and variable climate conditions. Under constant climate, mathematical modelling of viscoelasticity, thermo-viscoelasticity, and experimental results on relaxation of thin wood tissue and single fibres are reviewed. Under variable climatic conditions, modelling of transient hydrogen bonding effects on wood deformation, mechano-sorptive creep, and relaxation of single wood fibres and the influence of hemicelluloses on creep behaviour are presented.

Viscoelasticity of wood under constant climate

Under conditions of constant temperature and MC, the time dependent characteristics can be measured by static (creep, stress relaxation) and dynamic tests (Salmén and Hagen 2001). For characterization of linear viscoelastic behaviour, the creep and stress relaxation tests are generally preferred to dynamic tests. In static tests, the creep and relaxation function can be obtained explicitly on a time scale, whereas dynamic tests give a complex function in terms of frequency. The conversion of dynamic results and conclusion to static (creep function) is not always evident. Nevertheless, within the limits of linear viscoelasticity, it is demonstrated that the results obtained from these three methods are related mathematically.

The limit of the linearity domain of viscoelasticity should be established for defining the constitutive relations. This experimentally determined limit is related to wood species, the nature of loading, loading orientation with regard to the anisotropy, MC and temperature. Experiments have been mainly carried out in the longitudinal direction. Some of the results have been reviewed by Miller and George (1974) and Le Govic et al. (1990). These results indicate that the limit of linearity in bending along the longitudinal direction decreases with MC and is quite low being approximately 30% to 35% of the fail-

ure stress. However, in tension it is rather high (approximately 50%), while in compression, the stress limit is difficult to determine. Therefore, under moderate constant ambient climatic and moderate loading, the wood viscoelasticity may be considered linear (Whale 1988).

Characterisation of the linear behaviour of wood viscoelasticity

Orthotropic viscoelastic behaviour of wood is the result of its structural symmetry. The latest knowledge with regard to the structural features is summarised by Salmén and Burgert (2008). With the linearity assumption of viscoelasticity, the constitutive equation, which defines the relationship between time dependent strain (ε_{ij}) and stress (σ_{ij}) components, can be expressed in the time domain by the hereditary integral:

$$\varepsilon_{ij}(t) = \int_{0^-}^{t} S_{ijkl}(t-\tau)\frac{\partial \sigma_{kl}}{\partial \tau}d\tau \tag{1}$$

or

$$\sigma_{ij}(t) = \int_{0^-}^{t} R_{ijkl}(t-\tau)\frac{\partial \varepsilon_{kl}}{\partial \tau}d\tau \tag{2}$$

where, t is instance of observation, τ defines the time variable, S_{ijkl} and R_{ijkl}, stand, respectively, for the component of the fourth order creep compliance tensor S and relaxation tensor R.

Eqs. (1) and (2) represent Boltzmann's superposition integrals of a tri-axial iso-hygro-thermal stress-strain equation for a linear viscoelastic material.

In the case of plane orthotropic behaviour, the matrix representation of Eqs. (1) and (2) holds:

$$\begin{bmatrix}\varepsilon_{11}(t)\\ \varepsilon_{22}(t)\\ 2\varepsilon_{12}(t)\end{bmatrix} = \int_{0^-}^{t}\begin{bmatrix}S_{11}(t-\tau) & S_{12}(t-\tau) & 0\\ S_{12}(t-\tau) & S_{22}(t-\tau) & 0\\ 0 & 0 & S_{33}(t-\tau)\end{bmatrix} \frac{\partial}{\partial \tau}\begin{bmatrix}\sigma_{11}(t)\\ \sigma_{22}(t)\\ \sigma_{12}(t)\end{bmatrix} d\tau \tag{3}$$

$$\begin{bmatrix}\sigma_{11}(t)\\ \sigma_{22}(t)\\ \sigma_{12}(t)\end{bmatrix} = \int_{0^-}^{t}\begin{bmatrix}R_{11}(t-\tau) & R_{12}(t-\tau) & 0\\ R_{12}(t-\tau) & R_{22}(t-\tau) & 0\\ 0 & 0 & R_{33}(t-\tau)\end{bmatrix} \frac{\partial}{\partial \tau}\begin{bmatrix}\varepsilon_{11}(t)\\ \varepsilon_{22}(t)\\ 2\varepsilon_{12}(t)\end{bmatrix} d\tau \tag{4}$$

Hence, four scalar independent creep or relaxation functions have to be experimentally determined to characterise plane orthotropic time dependent behaviour of the wood. For a full characterising of the orthotropic time dependent behaviour, nine independent creep or relaxation functions are necessary. To determine these functions, laborious experimentations work should be performed (Schniewind and Barrett 1972; Cariou 1987; Hayashi et al. 1993).

In the literature, many mathematical expressions have been proposed to predict creep and stress relaxation functions. For linear viscoelastic behaviour, these models can be divided into two principal groups.

In the first group, after selecting a mathematical expression, the unknown coefficients are determined through creep curves by curve-fitting techniques. The typical mathematical expressions for creep functions are given, for example, by Navi and Heger (2005).

In the second group, the creep function is determined through a rheological model, made of series and parallel combinations of elastic springs and viscous dashpots by Bodig and Jane (1982), which properly represent the viscoelastic behaviour of the material. However, the unknown coefficients of the function are evaluated from the creep curve.

One of the rheological models commonly used for long-term viscoelastic behaviour of wood is the generalized Kelvin-Voigt model, presented in Figure 1. The corresponding time dependent function of creep is defined by:

$$S(t) = S_0 + \sum_{i=1}^{n} S_i\left(1-e^{-\frac{t}{\tau_i}}\right) + \frac{t}{\eta_\infty} \tag{5}$$

where, S_0, η_∞, S_i ($i=1,\ldots n$) and τ_l ($i=1,\ldots n$) are unknown coefficients.

Numerous mathematical expressions are suggested for creep and relaxation functions. One of the most common is the power law or parabolic model proposed by Nielsen (1984). The power law expression can be written as:

$$S(t) = S_0(1+(t/\tau)^k) \tag{6}$$

$$S_{\alpha\beta}(t) = S_{0,\alpha\beta}(1+(t/\tau)^k);\ \alpha = 1,\ldots 6 \text{ and } \beta = 1,\ldots 6 \tag{7}$$

Eq. (6) defines a creep function with three unknown coefficients (S_0, k, τ) for a one-dimensional medium, whereas Eq. (7) presents nine independent creep functions for a three-dimensional orthotropic material, where each function has three unknowns. All these unknowns are characterised through experimental creep tests. In these equations, τ appears as a constant called doubling time (where $S(\tau) = 2S_0$). Its value depends on the given MC of wood, temperature and the loading direction with regard to the principal axes of wood; S_0, $S_{0,\alpha,\beta}$ are the elastic compliance; and k is the factor of creep kinetics, which varies between $0 < k < 1$.

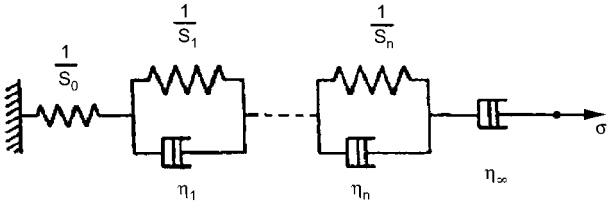


Figure 1 Elements of generalised Kelvin-Voigt model.

When stress level is high, wood may show non-linear viscoelastic behaviour. In this case, the creep and stress relaxation function becomes dependent on the loading stresses and, consequently, the creep or relaxation function loses its meaning. In this case, the relationship between time dependent strain and stress component in a one-dimensional medium may be described by the Green-Rivlin representation given in a summary by Morlier (1994):

$$\varepsilon(t) = \int_{0^-}^{t} S_1(t-\tau_1)\frac{\partial\sigma(\tau_1)}{\partial\tau_1}d\tau_1 + \int_{0^-}^{t}\int_{0^-}^{t} S_2(t-\tau_1,\, t-\tau_2)\frac{\partial\sigma(\tau_1)}{\partial\tau_1}\frac{\partial\sigma(\tau_2)}{\partial\tau_2}d\tau_1 d\tau_2 + \int_{0^-}^{t}\int_{0^-}^{t}\int_{0^-}^{t} S_3(t-\tau_1,\, t-\tau_2,\, t-\tau_3)\frac{\partial\sigma(\tau_1)}{\partial\tau_1}\frac{\partial\sigma(\tau_2)}{\partial\tau_2}\frac{\partial\sigma(\tau_3)}{\partial\tau_3}d\tau_1 d\tau_2 d\tau_3 + \ldots\ldots \quad (8)$$

In a test under a constant stress σ, the Green-Rivlin formulation becomes a non-linear polynomial function of stress and its third order reduced form presented in Eq. (9):

$$\varepsilon(t) = \sigma\, S_1(t) + \sigma^2\, S_2(t,\, t) + \sigma^3\, S_3(t,\, t,\, t) \quad (9)$$

In practice, it is usually found very difficult to calibrate the Green-Rivlin formulation by performing experiments even for Eq. (9). Other simplified alternative mathematical expressions for non-linear formulations are presented in the literature (Nakada 1960; Schapray 1966; Findley et al. 1976).

Stress relaxation in thin tissues and single wood fibres

A few experiments have been carried out on thin wood tissues by Kirbach et al. (1975) and Lofty et al. (1972) to investigate the influence of microfibril angle (MFA) on short-term creep behaviour. Recently, Eder et al. (2006) performed relaxation tests on tissue of 200 μm thickness and of single fibres of spruce (*Picea abies*) to study the time dependent behaviour of wood under constant temperature and humidity. The tissues and single fibres were prepared from both earlywood and compression wood. The fibres of these two parts had different microfibril angles. The thin wood tissues and corresponding single fibres were examined in order to separate mechanical phenomena occurring at the level of the tissue by means of cell-cell interaction from those occurring in the single fibre in the cell wall.

The results of the quoted authors indicate that relaxation is more pronounced for compression wood tissues and compression wood fibres than for tissues and fibres from earlywood. Since compression wood shows higher microfibril angle than early wood tissues, it was suggested that the relaxation behaviour of wood is more related to deformation inside the cell wall than to cell-cell interaction in thin tissue samples. The differences in relaxation of tissues and fibres within one tissue type may be explained by the critical role of cell geometry and the ability of the cell to undergo some torsion. Figure 2 demonstrates the relaxation curves of single wood fibres isolated mechanically from earlywood and compression wood. In Figure 2b and 2d, the forces were normalised to the initial instantaneous maximum force.

These results clearly show that fibres of compression wood are more viscoelastic than the fibres of normal wood, and it was concluded that these features are mainly determined by the different MFAs in the secondary cell wall 2 (S2 layer) of compression wood (~40–45°) and in the adult earlywood (~10–20°). However, apart from this, other structural and chemical features, such as cell length, cell diameter, shape, hemicellulose composition, lignin content and distribution, and the mechanical interaction of the polymers in the middle lamella are also considered to play a role. If hemicelluloses affect creep of wood, as attested by the authors in the successive part of this paper, it is not clear how high the contribution due to MFA is in comparison to the different composition in hemicelluloses.

Influence of temperature on viscoelasticity of wood

Wood is clearly exhibiting thermo-viscoelasticity (Huet et al. 1981; Jouve and Sales 1986; Salmén 1982) through creep and dynamical tests under different temperatures. Several models have been proposed to express wood creep dependence on temperature. One of the most suitable models for thermo-viscoelasticity is the combination of a power law expression, given in Eq. (10), with a thermo-activation law. This law concerns those polymers where principles of time temperature equivalency can be applied. The application of this principle to wood is still considered an assumption, which must be verified by experimentations.

The thermo-activation law has been defined in the form of an Arrhenius equation and also by the law of WLF (Williams, Landel and Ferry; Williams et al. 1955). The Arrhenius equation may be applied to temperatures below the glass transition temperature (T_g), while the law of WFL is more suitable for temperatures between T_g and $T_g + 50$°C (Salmén 1984; Salmén and Hagen 2001).

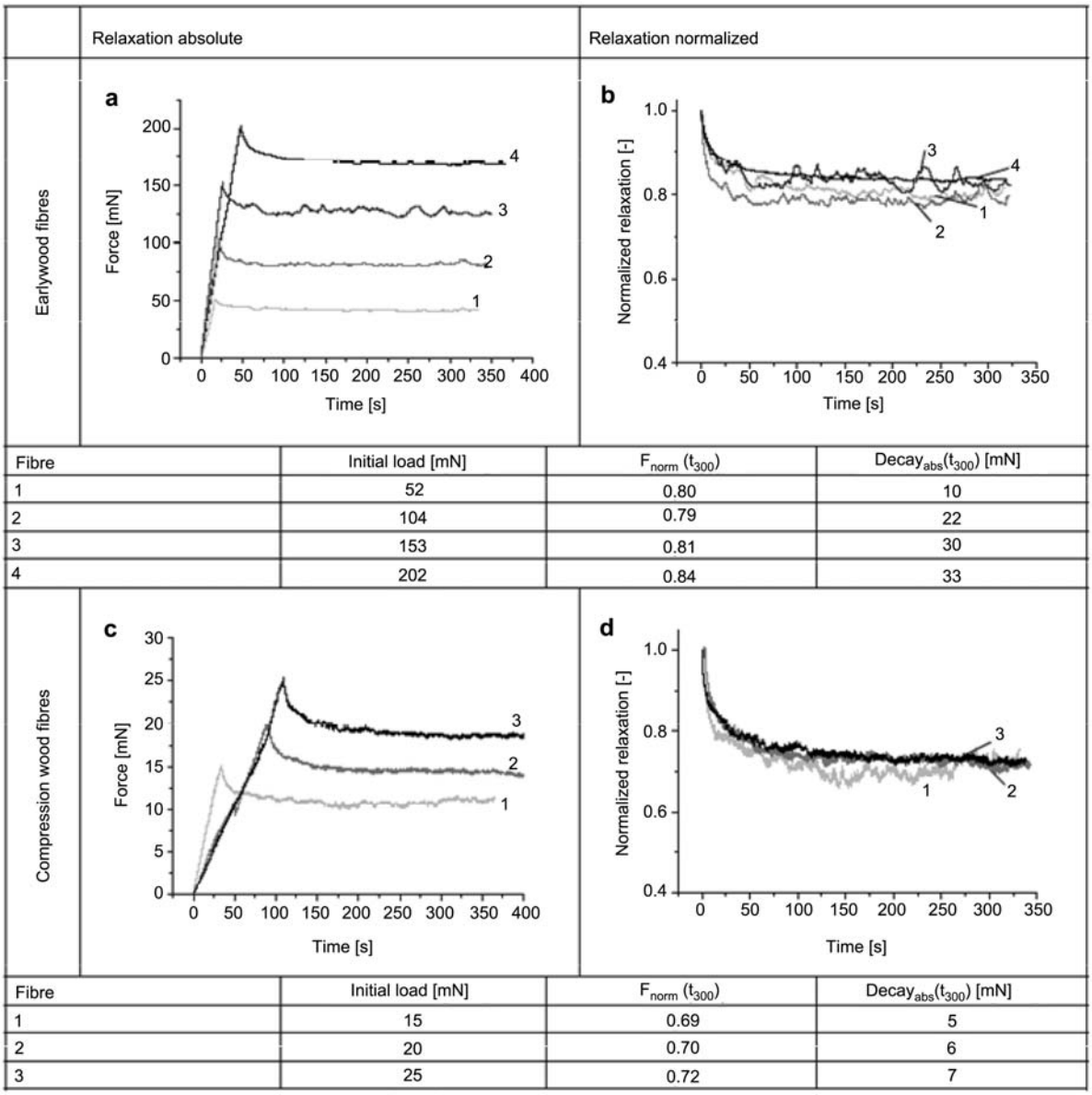
$$S(t) = S_0\,(1 + (t/\tau(T))^k) \quad (10)$$

$$\tau(T) = \tau_0\, e^{(W/R(T-T_\infty))} \quad (11)$$

Eq. (11) gives the dependency of τ on temperature in the form of the Arrhenius equation. In this equation, τ_0 is a constant, referring to infinite time, W is the activation energy (considered constant for each transition), R is the constant of a perfect gas.

Eq. (10) therefore includes the time and temperature dependence of the wood creep behaviour. This thermo-activation law has five parameters S_0, k, τ_0, W, T_∞ and two variables t and T.

Different experimental results (Poliszko 1986; Kauman et al. 1987; Salmén 1984; Le Govic et al. 1988) demonstrated that thermo-viscoelasticity of wood may involve



Fibre	Initial load [mN]	F_{norm} (t_{300})	$Decay_{abs}$(t_{300}) [mN]
1	52	0.80	10
2	104	0.79	22
3	153	0.81	30
4	202	0.84	33

Fibre	Initial load [mN]	F_{norm} (t_{300})	$Decay_{abs}$(t_{300}) [mN]
1	15	0.69	5
2	20	0.70	6
3	25	0.72	7

Figure 2 Representative relaxation curves of mechanically isolated fibres from earlywood and compressive wood; in (b) and (d) the forces are normalised by corresponding initial instantaneous maximum force (from Eder et al. 2006).

several second order transitions. Based on these results, Huet (1987) proposed a multi-transition model for wood thermo-viscoelasticity based on a complex compliance. This model consists of a serial link of parabolic elements (Huet 1987; Huet and Navi 1990). This type of modelling has shown a strong possibility in identifying all the parameters in the complex rheological model through experimental results.

Characterisation of the thermoactivated-viscoelastic behaviour of green wood has become essential for controlling the processes of wood transformation. This type of research is important regarding thermo-hygro-mechanical (THM) processing of wood. The influence of temperature on the viscoelastic behaviour of green wood in the transverse direction has been studied both by experimentation and modelling (Bardet 2001; Bardet et al. 2004; Vincent et al. 2004). During dynamic and static relaxation tests, the temperature was varied between 0°C and 100°C, while the wood samples were kept under saturated water conditions. The quoted authors observed that the mechanical properties of wood decrease drastically around 60°C. The results were presented in a complex plan showing the existence of two transitions. A multi-parabolic model based on Arrhenius law was employed and the parameters were determined through the experimental results. Simpler results were presented by Vincent et al. (2006) on the thermo-viscoelastic behaviour of poplar wood in the green state studied by torsional tests. They measured shear moduli (both loss and storage shear modulus) during relaxation tests using frequency sweep at different temperatures varying from 25°C to 75°C. In order to characterise the rheological model, the authors plotted the relaxation and dynamic results and the data were fitted by a portion of a circle. Based on these results, it was concluded that only one single parabolic model with three unknown parameters can represent the viscoelastic behaviour of green poplar in torsion. In a study, Tajvidi et al. (2006) examined the

effect of time temperature equivalency on the mechanical behaviour of a high-density composite made of kenaf fibre and polyethylene. The results indicated that this type of wood polymer composite was thermorheologically complex, and a single horizontal shift (probably equivalent to one transition) was not adequate to predict its long-term performances.

Viscoelasticity of wood under humidity variation

Many experiments have shown that a cyclic variation of the environment increases the creep of wood and wood based products (Armstrong and Kingston 1960; Pittet 1996; Ebrahimzadeh and Mcqeen 1998; Valentin and Chaplain 2002; Chaplin 2006). This phenomenon is often referred to as mechano-sorptive creep (Grossman 1975) and it has direct effects on fracture, time dependent deformation and stress relaxation of wood in structural applications, cardboard boxes, paper, paper-based packing materials and wood-reconstituted panels. One of the undesired effects of mechano-sorptive creep is that delayed failures can occur at relatively low loads and short periods of time upon cycling of the relative humidity.

The same phenomenon has been demonstrated in other hygroscopic materials, such as cellophane, Kevlar 29, polyethylene oxide and other polymers (Wang et al. 1990, 1991; Ebrahimzadeh and Kubàt 1997).

Since the pioneering work of Armstrong and Kingston (1960), numerous investigators studied the origin of this effect (Gibson 1965; Hunt 1984, 1986; Gril 1988), whose contribution has been reviewed by Wang et al. (1991). However, the physical mechanisms governing the mechano-sorptive effects remain poorly understood (Hunt and Gril 1996; Hankijäwi and Hunt 1998; Navi et al. 2002; Randriambololona 2003). In the following, new results are illustrated and discussed concerning the behaviour of thin wood strips and single wood fibres in the context of the role of hemicelluloses and micromechanics of transient moisture effects on wood creep.

Role of hemicelluloses on creep behaviour of wood

There is some evidence that the hemicelluloses are mostly responsible for the time dependent behaviour and mechano-sorption effects of wood. Experiments addressing this issue were carried out by Fioravanti et al. (2006). Two sets of clear samples of spruce (*Picea abies*) were investigated: the first set being natural wood as a reference, while in the second set the hemicelluloses were removed. It is difficult to confirm the complete removal of hemicelluloses, although it has been proven by IR spectrometry based on the xylan absorption at the wave number 1740 cm^{-1}. Experiments were performed at 20°C, while relative humidity (RH) was cycled from 30% to 75% during application of the force. Applied loads were 20% and 40% of ultimate tensile strength, respectively. The results are given in Figure 3.

The main results confirm that: (1) at the same RH%, the samples without hemicelluloses show different MC at equilibrium than natural wood; (2) the time to reach MC equilibrium is longer for samples without hemicelluloses, these samples show a lower viscoelasticity than natural wood under constant climatic conditions; and (3) during RH variations, the mechano-sorptive creep of natural wood has accelerated more than wood without hemicelluloses. The same results also indicate that wood without hemicelluloses still shows viscoelasticity and mechano-sorption creep but to a lesser degree.

Mechano-sorptive creep of single wood fibres

Creep tests were carried out under controlled cycling humidity both on single wood fibres and pulp fibres by Salmén et al. (2006) to verify that single fibres experience

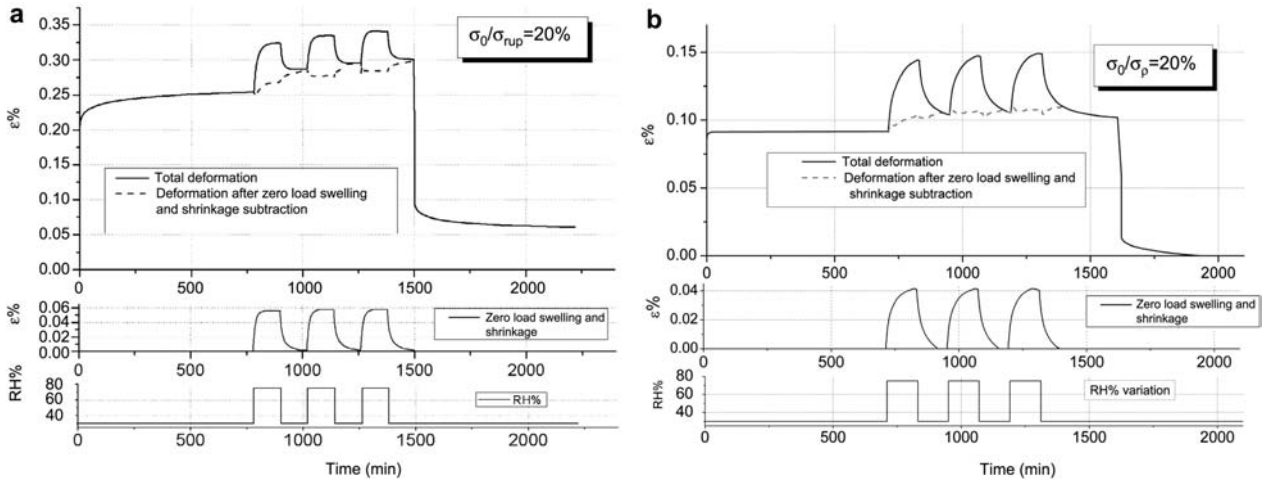


Figure 3 Examples of creep-recovery tests on thin spruce tissue of 0.7 mm thick in tangential direction under tensile loading of 20% of rupture force at 20°C. The first part is from 0 to 750 min under a constant humidity of 30% RH and the second phase from 750 to 1500 min under a three-cyclic humidity at 30 and 75% RH. At 1500 min the specimens were totally unloaded; (a) test on natural spruce, (b) test on spruce samples without hemicelluloses (from Fioravanti et al. 2006).

mechano-sorptive creep. Fibres were isolated mechanically to retain the natural constituents of the secondary wall (Burgert et al. 2005), while pulp fibres were separated chemically by maceration of spruce chips by means of a mixture of hydrogen peroxide and acetic acid. Single fibres tested in a Perkin Elmer dynamic mechanical analyser at 30°C fixed temperature clearly showed an increased creep in cyclic humidity (30% to 80% RH) as compared to the creep at constant humidity (80% RH).

Examples of creep curves for a wood and a pulp fibre are given in Figure 4 in a logarithmic time scale. In Figure 4, strain has been set to zero at a loading time of 1 min, ignoring the instantaneous elastic strain. In this example, the load is higher for the wood fibre than for the pulp fibre.

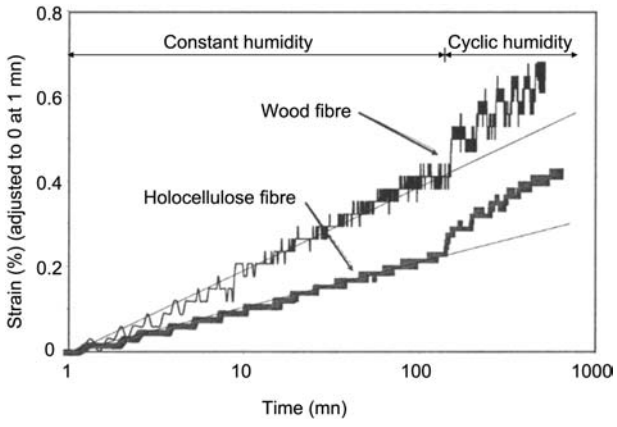


Figure 4 Creep curves on a logarithmic time scale for a single wood fibre and a single pulp fibre at 30°C, respectively; the first phase is from 0 to 120 min under a constant humidity of 80% RH and the second phase from 120 min to 615 min under cyclic humidity at 30% and 80% RH; each period was of 30 min duration (from Salmén et al. 2006).

Mechano-sorptive relaxation of single wood fibres

Stress relaxation behaviour of single wood fibres under humidity cycling was studied by Navi et al. (2006). The fibres were separated mechanically (Burgert et al. 2005). A major difficulty in manipulating the single fibres is their tendency to twist during drying. Therefore, the fibres were microscopically observed and highly twisted fibres were eliminated from the relaxation tests. The tests were performed in a special tensile testing machine designed and constructed for this purpose.

To estimate the stress level during mechano-sorption tests, many force-strain tests were carried out under various constant temperatures and RH. In all the experiments, displacements were applied manually by a deformation screw at a ratio of 5 μm per 5 or 6 s. Figure 5 illustrates a typical force-strain curve of a single fibre under tensile force where two loadings and one unloading were performed.

Figure 5 shows that the first segment of the curve from A to B corresponds to straightening of the fibre in the load direction. For each fibre, this segment can vary. The second segment from B to C is where the force increases

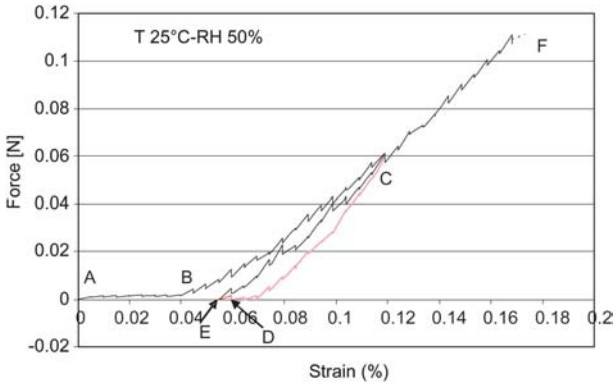


Figure 5 Typical force-strain curve for a single wood fibre of spruce under loading-unloading- and loading; temperature and RH were kept constant during the test, displacements were applied manually by a deformation screw at ratios of 5 μm per 5 or 6 s (from Navi et al. 2006).

with increasing deformation where the slope indicates an almost linear stress-strain behaviour. At point C, the displacement of the fibre was decreased until point D where the stress in the fibre approached zero. The unloaded curve does not take the same path as the loading curve. This is mainly due to the viscoelasticity of the wood cell. At point D the force tends to increase while the displacement is kept constant. This phenomenon is called anti-relaxation. This could be visualised clearly if the strain and the corresponding stress curve were plotted on a time scale. The displacement was again decreased to a point E where the stress returned to zero. The displacement between the two points E and B is most probably due to fibre plasticity behaviour under loading until point C on the curve. Several authors (Navi et al. 1995; Keckes et al. 2003) have already mentioned this type of plastic deformation of thin wood specimens under tensile loading at its longitudinal direction.

It is realised from force-strain experiments that under forces ranging between 20 and 40 mN the behaviour of fibres remain linear. Temperatures between 20°C and 40°C have a small effect on the behaviour of the fibre, but the rupture stress decreased sensibly for temperatures higher than 40°C. Therefore, relaxation tests under humidity variation were carried out at temperatures between 20°C and 40°C and forces in the range of 20 to 40 mN. A large number of tests were performed to investigate the mechano-sorptive relaxation of single wood fibres (Meylan 2006). One should note that during relaxation tests it was required to ensure good alignment of the fibre in the force direction and performing the test at the fibre linear stress-strain range. An example of relaxation curves of a single wood fibre under cyclic humidity is given in Figure 6.

One can verify that during cycling of humidity the fibre reveal increased relaxation with time. Especially, one should note that relaxation is increased each time if the fibre undergoes drying. Therefore, both the creep curve illustrated in Figure 4 and the stress relaxation behaviour given in Figure 6 clearly demonstrate that single wood fibres present mechano-sorptive creep and mechano-sorptive relaxation, respectively. Thus, earlier claims by Sedlachek et al. (1994) and Sedlachek (1995), that single

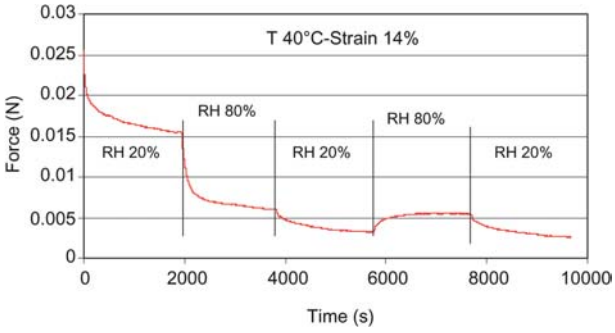


Figure 6 Relaxation curve of a single wood fibre under variable humidity and constant temperature; the first segment is from 0 to 2000 s at a constant 20% RH; the second segment from 2000 to approximately 10 000 s under two humidity cycles from 80% to 20% RH; each cycle was a 4000-s period. Drying part of the humidity cycles always accelerates the relaxation and re-humidification part increases internal forces (here it is called anti-relaxation) except on the first cycle. This curve illustrates the total load without compensation for loads due to swelling and shrinkage (from Navi et al. 2006).

wood fibre does not experience mechano-sorptive creep cannot be generalised.

Modelling the effect of transient hydrogen bonding at molecular level

It was demonstrated that mechano-sorption is a complex phenomenon which is ascribed to transient hydrogen bonding, changing in molecular mobility, and internal stress gradients due to the presence of moisture gradients. However, the underlying mechanisms remain poorly understood.

To explore this phenomenon, Navi et al. (2002, 2005) have numerically modelled the effect of continuous breaking and reforming of hydrogen bonds at the wood molecular level through the movement of water molecules, based on a concept which was first proposed by Gibson (1965). In this model, the structure of wood is idealised as a periodic fibre-reinforced composite, whose reinforcing elements were extended chain crystals of cellulose embedded in a matrix of amorphous cellulose considered to represent the hemicelluloses. This composite model comprises alternating layers of cellulose and amorphous cellulose.

The super cell used in the simulation was constructed from 24 unit cells of cellulose Iα and an equivalent number of "amorphous" sequences, which were lightly cross-linked to prevent crystallisation during subsequent relaxations.

Dynamics simulations were carried out by the Cerius 2 simulation software package (Molecular Simulation Inc.). Periodic boundary conditions were applied and the structure minimised and relaxed at 350 K and at constant pressure for 100 ps (picoseconds) prior to application of a stress. Where required, the water (5–10 wt% overall) could be made to diffuse in and out of the super cell by placing a sink at both ends of the amorphous layer (water was found not to enter or bond to the interior of the crystalline phase in significant amounts). However, even given the reduced size of the model, this took too long for a systematic parametric study of the coupling between diffusion and mechanical deformation to be contemplated in the time available (under diffusion control, the size of the sample ought to be an important factor). The aim has rather been to demonstrate coupling between moisture and the mechanical response of the system. Therefore, the investigation was restricted to a temperature of 350 K and to relatively large shear stresses acting parallel to the chain direction in the plane of the layers, such that the effective slip planes are perpendicular to the hydrogen bonding direction in the crystallites.

Figure 7 illustrates the effect of applying a 200-MPa shear force to the model while allowing water to diffuse out of the super cell. The water molecules were initially numerous at the amorphous-crystalline interface, participating in the hydrogen bonding between the two phases, and remained almost entirely segregated to the

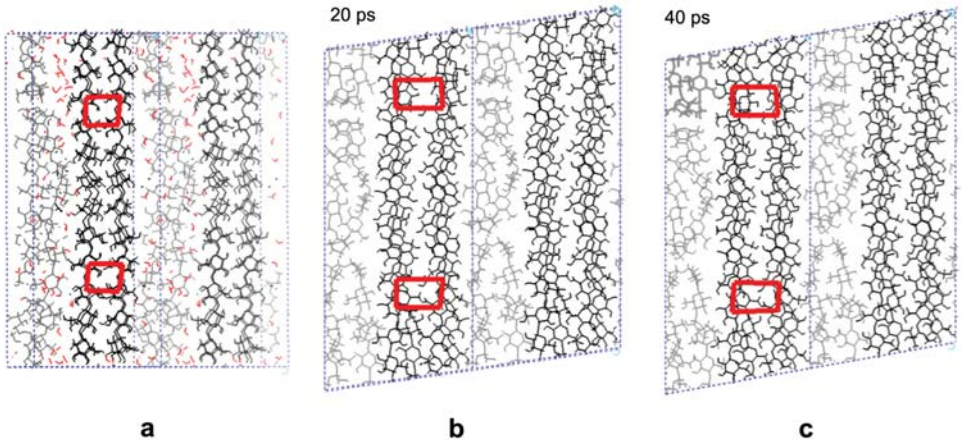


Figure 7 Effect of coupling between the shear force and drying of the model. Input values were: shear stress 200 MPa acting parallel to chain direction in the plane of the layers; temperature 350 K and initial water content 10% (by wt). (a) Composite model with periodic boundary composing alternating layers of cellulose and amorphous cellulose at initial state; (b) effect of applying shear stress while allowing water to diffuse out during 20 ps; (c) shear deformation after 40 ps. Shear strain of crystalline phase (the rectangles plotted in crystalline cellulose remain at right angle before and after application of shear force) is very small compared to global shear strain (from Navi et al. 2002).

amorphous phase throughout the simulation. Some of the initially hydrogen-bonded water molecules are able to diffuse out the super cell over the time-scale of the simulation and others undergo substantial displacements within the amorphous phase (unbound water molecules diffuse rapidly as verified by turning off atomic charges during certain runs). The shear strains in the crystalline phase are very small compared with the global strains, indicating slip in the amorphous phase or at the interface to dominate the shear strain. In particular, both the intra- and inter-molecular hydrogen bonds in the crystalline phase remain intact, even under very large external shear stresses. A series of simulations of this type indicated the deformation rates to be enhanced with respect to those in relaxed models containing either no water or to models in which the water content is maintained constant. We suggested based on the results that transient hydrogen bonding in the amorphous phase and at the interface between the crystalline cellulose and hemicelluloses may accelerate shear slip between two phases in the presence of an external load due to the introduction or removal of water molecules.

Conclusion

A micromechanical approach has become an important tool for verifying and suggesting deformation models on various structural levels of materials. The aim is a better prediction of the deformation at micro-structural level, to optimise macroscopical behaviour and to explain phenomena, such as the damage and fracture processing, the scale effects, thermo-viscoelasticity, apparent plasticity, the hygro-expansion and transient phenomenon in materials. A micromechanical approach can have various stages: experimentations, observations and modelling.

A micromechanical approach to wood resulted in new results on viscoelastic behaviour of wood in recent years. Some new issues reviewed in this article are:

- The origin of stress relaxation of wood is complex but more related to phenomenon intervening inside the cell wall than cell-cell interaction in thin tissue. On the other hand, the origin of stress relaxation of a single wood cell mainly related to the critical role of cell geometry and the ability of the cell to undergo torsion.
- Hemicelluloses play a very active role on the viscoelastic behaviour and moisture adsorption of wood. Wood samples with removed hemicelluloses adsorb less MC at equilibrium, time to MC equilibrium is longer, and viscoelasticity and mechano-sorption is lower than for natural wood. However, the samples without hemicelluloses still exhibit viscoelasticity and mechano-sorption creep but in lesser amounts.
- Single wood fibres have mechano-sorptive creep and mechano-sorptive relaxation. Thus, earlier claims that single wood fibre do not show this behaviour cannot be generalised. These results confirm that the origin of the mechano-sorptive effect is located inside the wood cell wall.
- The results of numerical modelling the effect of continuous breaking and reforming of hydrogen bonds at the molecular level concerning the movement of water molecules can be interpreted that transient hydrogen bonding in the amorphous (paracrystalline) phase of cellulose and at the interface between the crystalline cellulose and hemicelluloses may accelerate shear slip between phases in the presence of an external load due to the introduction or removal of water molecules. This result confirms the concept which was first proposed by Gibson (1965).

The polymeric components of wood and their unique multilevel hierarchical arrangement confer to wood a versatile character and a possibility for THM transformation exceeding that of all other materials. The thermo-hygro-viscoelasticity (THV) properties of wood are involved in numerous transformation processes under action of THM, such as bending, thermo-shaping, densification, heat treatment for dimensional stabilisation, welding, wet wood heating for dissipating the internal energy and machining. To improve THM transformation processes it is necessary to better understand the THV properties of wood and develop corresponding modelling.

The understanding and modelling of THV behaviour of wood is difficult because of its polymeric nature consisting of hydrophilic cellulose and hemicelluloses and hydrophobic lignin and because hydrogen bonds and van der Waals forces are active between them. Further understanding of THV of wood – including its mechano-sorption effects and multi-thermoactivation – requires studies on the role of the hemicelluloses and lignins at viscoelasticity, precise determination of the supramolecular structure of the cell wall and the nature of the reversible bonding among its constituents. Some insight into this field may be found in a review by Salmén and Burgert (2008).

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